Thermal Aging Embrittlement of High Chromium Oxide Dispersion Strengthened Steels

J.S. Lee[1)], I.S. Kim[1)], C.H. Jang[1)], A. Kimura[2)], B.G. Kim[3)], K.N. Choo[3)], Y.S. Choo[3)], Y.H. Kang[3)]
1) Department of Nuclear and Quantum Engineering, KAIST 373-1, Daejeon 305-701, Korea, jslee2@kaist.ac.kr
2) Institute of Advanced Energy, Kyoto University, Gokasho, Uji, Kyoto, Japan, kimura@iae.kyoto-u.ac.jp
3) Korea Atomic Energy Research Institute, P. O. Box 105, Daejeon 305-600, South Korea

1. Introduction

Oxide dispersion strengthened (ODS) steels are considered as one of the promising candidate materials not only for fusion reactor blanket system, but for fuel clad materials of high burn-up operation of light water reactor (LWR) and supercritical pressurized water reactor (SCPWR).

Recent experimental works indicated that the ODS steels were highly resistant to irradiation embrittlement up to 15 dpa [1]. The high Cr ODS steels whose Cr concentration were in the range of 15 to 19 wt. % showed high resistance to corrosion in SCPW [2]. As for the hydrogen embrittlement, the critical hydrogen concentration required for brittle cracking was about 10~12 wppm that is approximately one order higher value than that of 9Cr reduced activation martensitic steels [3]. However, there were limited data for thermal aging embrittlement of high Cr ODS steels at temperatures from 673 to 773 K. In general, aging of high Cr steels for extended periods at temperatures between 673 and 823 K can result in the precipitation of coherent particles of ά (Cr-rich ferrite) with an associated reduction in ductility [4].

In this work, the effects of thermal aging treatment on the microstructural stability and mechanical property changes of ODS steels were investigated by using TEM, microhardness and small punch (SP) tests.

2. Experimental

The materials used were five kinds of ODS steels produced by varying Cr content from 13 to 22 wt. % but keeping yttria contents within 0.36~0.38 wt. %. The ODS steels were indexed as K1 to K5. Disk type SP specimens ($3\varphi \times 0.28^t$ mm) were sampled from the extruded rod so that the axis direction is parallel to longitudinal (L) or transverse (T)-direction with respect to the extruded direction. Specimens were thermally aged at 693 K up to 322 hours, which corresponded to the 50 months operation as a fuel clad in LWR. Thin foils for transmission electron microscopy (TEM) were fabricated by twin jet electropolisher in a solution of 10% perchloric acid + 90% ethanol at -30 ^oC. SP tests were performed at a cross-head speed of 0.2 mm/min. at temperatures from 303 to 77 K. SP ductile to brittle transition temperature (SP-DBTT) was defined as a temperature where the SP energy was the average of maximum and lower shelf SP energy. Vickers micro-hardness test was carried out with a 500 g load at room temperature.

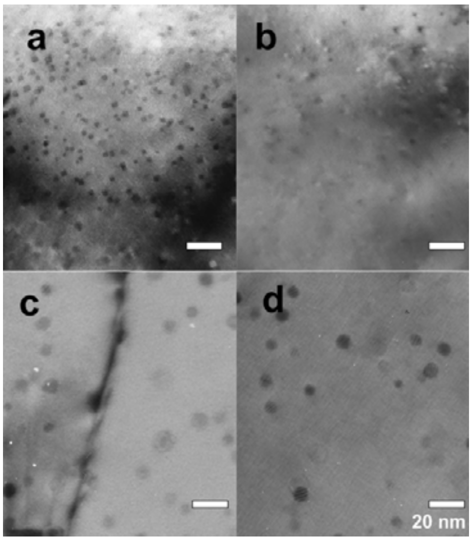

Figure 1. Typical transmission electron micrographs of K1 and K5 ODS steel, showing as received (a) K1, (c) K5 and thermally aged (b) K1, (d) K5.

3. Results

3.1. Microstructural characteristics

Typical microstructure of K1 (19Cr) and K5 (22Cr-4Al) ODS steels is shown in Figure 1. The average diameter and number density of yttria particles in K1 ODS steel was measured 2.4 nm and 1.4×10^{23} m^{-3}, respectively (Figure 1a). However, when 4 percent Al was added, the size and number density of yttria changed from 2.4 to 6~7 nm and from 1.4×10^{23} to $1\sim2\times10^{22}$ m^{-3}, respectively, regardless of Cr content. Detailed information was summarized in Table 1. After thermal aging treatment at 693 K for 322 hours, any discernable precipitation could not be found in the ODS steels (Figure 1b, d). Considering the results of SP and microhardness tests, however, very fine (<1 nm) precipitations must had been formed.

Table 1. Size and number density of yttria particles in each ODS steel before and after thermal aging treatment.

	As-received			Thermally aged		
	Size (nm)	Density (m^{-3})	Mean distance (nm)	Size (nm)	Density (m^{-3})	Mean distance (nm)
K1 (19Cr)	2.4	1.4×10^{23}	55	-	-	-
K2 (13Cr-4Al)	6.3	2.3×10^{22}	83	6.6	2.2×10^{22}	83
K3 (16Cr-4Al)	6.8	1.4×10^{22}	102	6.6	1.6×10^{22}	97
K4 (19Cr-4Al)	6.7	1.6×10^{22}	97	8.8	1.4×10^{22}	90
K5 (22Cr-4Al)	7.3	1.3×10^{22}	103	7.7	1.3×10^{22}	100

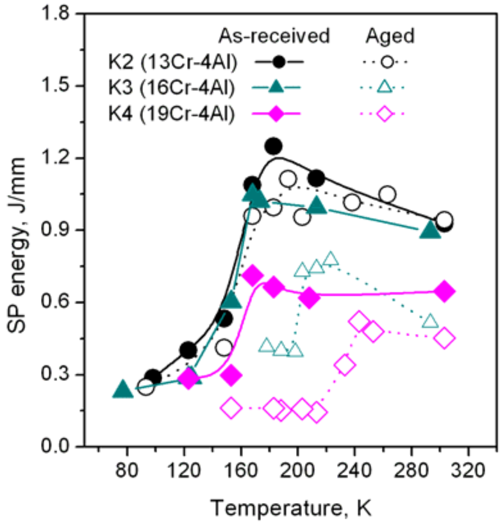

Figure 2. Effects of thermal aging treatment on the specific SP energy of ODS steels as a function of test temperature in T-direction, thermally aged at 693 K for 322 hours.

3.2. SP ductile to brittle transition behavior

The changes in SP energy by thermal aging are summarized in Figure 2, showing an increase in SP-DBTT and reduction in upper shelf energy. In as-received K2, K3 and K4 T ODS steel, it showed an increase in SP energy with decreasing test temperature up to 160 K, and abrupt drop of SP energy was observed by further cooling. As expected from the tensile properties, K1-T ODS steel revealed the lowest SP energy at all temperatures, and the SP-DBTT was about 170 K, irrespective of the ODS materials. Furthermore, the ductile to brittle transition behavior was strongly dependent on the specimen sampling direction, namely,

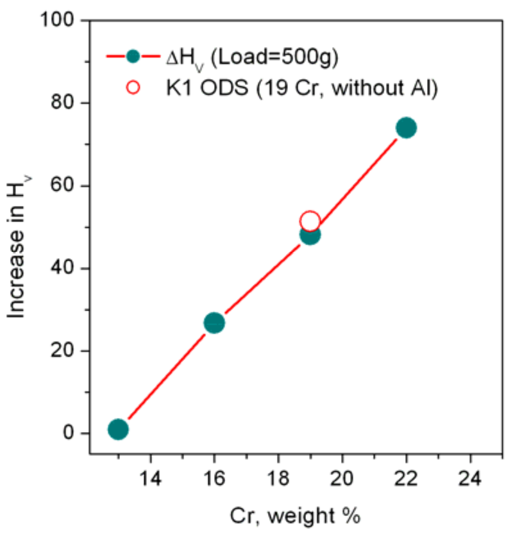

Figure 3. Increase in Vickers micro-hardness as a function of Cr content in ODS steels. (Load=500g).

L- or T- direction. Generally in L-direction showed lower SP-DBTT values than those of T-direction.

The degree of aging embrittlement increases with Cr content such that the increase in SP-DBTT associated with the reduction in upper shelf energy is 10, 50 and 73 K in 13 (K2), 16 (K3) and 19 Cr (K4) ODS steel, respectively. Figure 3 shows the hardening of ODS steels due to thermal aging as a function of Cr content. As the Cr content increased from 13 to 22 w/o, the increase in hardness (ΔHv) ranged from 1 to 74. However, there was no effect of Al addition on the total amount of hardening. Based on the TEM investigation, the origin of hardening must be come from the invisibly fine size of Cr-rich ά-phase formed by nucleation and growth mechanism in Fe-Cr system.

4. Summary

After thermal aging treatment at 693 K for 322 hours, any discernable precipitation could not be found in the 13~22 Cr ODS steels, but showing an increase in SP-DBTT and reduction in upper shelf energy. The degree of aging embrittlement increases with Cr content such that the increases in SP-DBTT are 10, 50 and 73 K in 13, 16 and 19 Cr ODS steel, respectively. Amount of hardening also increased linearly as a function of Cr content. Based on the TEM observation, the origin of hardening must be related to the invisibly fine size of Cr-rich ά-phase (<1 nm).

Acknowledgment

This work was partially supported by the Ministry of Science and Technology of Korea through the HANARO Utilization Program which is one of the National Nuclear R&D Program (2004).

REFERENCES

[1] T. Yoshitake, T. Ohmori and S. Miyakawa, J. Nucl. Mater. 307-311 (2002) 788.
[2] H.S. Cho, A. Kimura, S. Ukai, M. Fujiwara, J. Nucl. Mater. 329-333 (2004) 387-391.
[3] J.S. Lee, A. Kimura, S. Ukai, M. Fujiwara, J. Nucl. Mater. 329-333 (2004) 1122-1126.
[4] R. L. Klueh, D.R. Harries, High-Chromium Ferritic and Martensitic Steels for Nuclear Applications, ASTM, 2001, p.39-55.